LEAD SOIL TREND ANALYSIS THROUGH JULY 2007 EVALUATION BY INDIVIDUAL QUADRANT

HERCULANEUM LEAD SMELTER HERCULANEUM, MISSOURI

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INTRODUCTION

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Tetra Tech EM Inc. (Tetra Tech) was tasked by the U.S. Environmental Protection Agency (EPA) Region 7 Enforcement/Fund Lead Removal program to conduct a trend analysis of soil lead concentrations at selected locations within Herculaneum, Missouri (City). Specifically, the Tetra Tech Superfund Technical Assessment and Response Team (START) 2 was requested to review and analyze data that would enable EPA to determine if soil lead concentrations were increasing over time at a variety of locations within the City. Two tasks were identified: (1) perform a trend analysis for individual quadrants within each yard using the most current sampling data, and (2) estimate the range of monthly increase in lead concentrations for properties grouped into three categories based on distance from the smelter (less than or equal to 025 mile, 0.25 to 0.50 mile, and 0.50 to 0.75 mile). The assessment was conducted under the authority of the Comprehensive Environmental Response, Compensation, and Liability Act of 1980 and the Superfund Amendments and Reauthorization Act of 1986. The project was assigned under START Contract No. 68-S7-01-41, Task Order No. 0021.

Tetra Tech focused its analysis on one data set called "Recontamination." This data set includes results from a number of residential properties. The data were collected from four different quadrants at each property, and additional data for several properties came from samples collected in driveway areas outside the quadrants. Lead sampling was conducted at each location at varying intervals from the time removal activities were completed in early 2002 (sampling round 6). Sampling was conducted monthly prior to 2003, quarterly from 2003 to 2004, and semi-annually after October 2005 (sampling round 22). This report includes results for sampling conducted between August 2002 (sampling round 7) and July 2007 (sampling round 25). Due to the sequence of removal activities, not all properties underwent the same number of sampling events; the number of events ranged from 6 to 19 events per quadrant for individual properties. At many locations, some intervals within the series were omitted because of weather or access restrictions. The lead concentrations were determined by use of a portable X-ray fluorescence (XRF) instrument. Samples were collected and analyzed in accordance with the quality assurance project plan (QAPP) dated September 11, 2001.

This document presents the methods used to evaluate changes in soil lead concentrations following the removal activities, and the results of this analysis.

METHODS

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Trend tests were conducted for each property using data collected from round 7 (August 2002) through round 25 (July 2007). The non-parametric Mann-Kendall test was used to evaluate temporal trends for each sampled quadrant at the individual properties. The Mann-Kendall test is a widely used statistical test for detecting monotonic trends (that is, trends either increasing or decreasing) in time-series of data (Gilbert 1987; Helsel and Hirsch 1992; Gibbons 1994). Because the Mann-Kendall test uses only the relative magnitude of the data rather than their measured values, it has a number of desirable properties: the data need not be normally distributed; and the test is not significantly affected by outliers, missing data, or censored data. Censored data are treated in the Mann-Kendall test by setting all nondetect values to a concentration slightly below the minimum detected concentration. It should be noted that a minimum of four sampling events are required to perform this test, so properties with fewer than four rounds of sampling were not evaluated. Properties not sampled during round 25 were also excluded from the trend analysis.

For all properties where at least one quadrant showed a significant increasing trend based on the Mann-Kendall test, regression analysis was performed to estimate the monthly increase in lead concentration. This analysis was performed to provide rough estimates of the range of potential increase in lead concentrations for properties grouped according to distance from the smelter. Three distance categories were evaluated: less than or equal to 0.25 mile, 0.25 to 0.50 mile, and 0.50 to 0.75 mile. Because the purpose of this analysis was to only provide rough estimates of the rate of change in lead concentration, regression was performed on the data in original units (i.e., untransformed data). It should be noted that certain evaluation methods and diagnostic tools commonly used in linear regression analysis (e.g., evaluation of different transformations of the data, verification of model assumptions, and evaluation of outliers) were not used in this analysis.

For quadrants with detected data only, ordinary least squares (OLS) linear regression analysis was used. For quadrants with one or more censored (nondetect or ND) measurements, a censored maximum likelihood estimation (MLE) approach was used, following Helsel (2005). Censored MLE methods are increasingly used in environmental assessment work, given the increased speed of modern personal computers and the enhanced capabilities that have been added into many commercial statistical software packages. As described in Helsel (2005), MLE regression techniques can be implemented using commercial software with capabilities for performing parametric survival analysis on interval-censored

data. It should be noted that MLE regression for left-censored data is also referred to as "Tobit analysis" in the technical literature. MLE methods recognize each censored datum as an interval, bounded by zero at the lower limit and the detection or reporting limit at the upper limit. Application of OLS regression with censored data is contraindicated, as it requires substitution of an assumed value (typically zero, the detection limit, or one half the detection limit) for each censored datum, resulting in biased estimates for the regression parameters.

RESULTS

Temporal trends in lead concentrations for 14 properties are summarized in Table 1 and Figure 1. The trend analysis identified 11 out of 14 properties where at least one quadrant showed a statistically significant increasing trend. No statistically significant decreasing trends were identified for any properties. Six properties had increasing lead concentrations in all four quadrants: house numbers 5, 9, 18, 19, 22, and 24. Two properties had increasing lead concentrations in three of four quadrants: house numbers 3 and 6. Two properties had increasing lead concentrations in two of four quadrants: house numbers 7 and 76 (only two quadrants evaluated). House number 13 had increasing lead concentrations in one quadrant. Three properties—house numbers 10, 103, and 104—showed no statistically significant trend in lead concentrations in any quadrant. All trend results are depicted graphically in Figure 1. Open symbols are used in Figure 1 to represent censored (nondetect) data, and solid symbols represent detected data.

Trend results reported for soil lead concentrations through sampling round 25 were similar to those reported during the last quarterly period. No changes in trend were seen for the same subset of houses and quadrants sampled in both rounds 24 and 25. Note that houses 20 and 101 were sampled during round 24 but were not sampled during round 25; houses 10, 13, and 19 were sampled in round 25 but not in round 24.

Table 2 shows the results of OLS and MLE regression analysis performed on properties that showed a significant increasing trend in lead concentration in at least one quadrant. The slope, intercept, standard error of the slope, and two-sided 95 percent confidence intervals for the slope estimates were calculated for 33 quadrants within 10 properties. Ranges for the monthly rates of increase in lead were 0.66 to 8.37 milligrams per kilogram per month (mg/kg/month) for properties located less than or equal to 0.25 mile from the smelter; 0.81 to 3.53 mg/kg/month for properties located between 0.25 and 0.50 mile from

the smelter; and 0.90 to 6.45 mg/kg/month for properties located between 0.50 to 0.75 mile from the smelter. The upper 95 percent confidence limit (UCL) for the monthly rate of increase was also evaluated to estimate maximum potential rates of increase. Because of the variability in the individual estimates, the 50th, 75th, and 90th percentiles of the distribution of the individual UCLs within each distance category are also reported in Table 2. The 75th and 90th (in parentheses) percentile values for the monthly rate of increase for the properties grouped according to increasing distance from the smelter are 4.88 (7.64), 3.72 (5.04), and 3.40 (9.03) mg/kg/month. It should be cautioned that these are considered rough estimates only, as no attempt was made to evaluate the validity of the regression model assumptions, or the uncertainty associated with the predicted rates of increase.

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- Helsel, D. 2005. Nondetects and Data Analysis: Statistics for Censored Environmental Data. John Wiley & Sons, Inc. New York, NY. 250 pages.
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TABLE 1

RESULTS OF STATISTICAL TESTING FOR MONOTONIC TRENDS (MANN-KENDALL TEST) IN LEAD CONCENTRATION INDIVIDUAL QUADRANTS FOR SAMPLING ROUNDS 7 THROUGH 25 HERCULANEUM LEAD SMELTER SITE — HERCULANEUM, MISSOURI

Distance			Number of	Number of	Sampling Event		Mann-		Trend	
From	House		Sampling	Detected		THE PAR	Kendall Test		Significant? ⁴	Direction of
Smelter ¹	Number	Quadrant	Events ²	Samples	First	Last	Statistic ³ (S)	> S	(Yes/No)	Trend
0.10	76	Q1	12	12	10/30/2003	07/10/2007	50	0.001	Yes	Increase
0.10	,,,	Q2	12	12	10/30/2003	07/10/2007	38	0.008	Yes	Increase
		Q1	18	15	08/26/2002	07/11/2007	121	< 0.001	Yes	Increase
	5	Q2	18	17	08/26/2002	07/11/2007	123	< 0.001	Yes	Increase
		Q3	18	18	08/26/2002	07/11/2007	108	< 0.001	Yes	Increase
		Q4	18	18	08/26/2002	07/11/2007	105	< 0.001	Yes	Increase
	6	Q1	18	18	08/23/2002	07/11/2007	47	0.044	Yes	Increase
0.25		Q2	18	18	08/23/2002	07/11/2007	64	0.012	Yes	Increase
		Q3	18	18	08/23/2002	07/11/2007	8	0.193	No	N/A
		Q4	18	18	08/23/2002	07/11/2007	53	0.029	Yes	Increase
		Q1	17	17	08/26/2002	07/10/2007	54	0.018	Yes	Increase
	22	Q2	17	17	08/26/2002	07/10/2007	68	0.004	Yes	Increase
		Q3	17	17	08/26/2002	07/10/2007	89	< 0.001	Yes	Increase
		Q4	17	17	08/26/2002	07/10/2007	86	< 0.001	Yes	Increase
		Q1	15	15	11/07/2002	07/10/2007	57	0.004	Yes	Increase
	24	Q2	15	15	11/07/2002	07/10/2007	75	< 0.001	Yes	Increase
		Q3	15	15	11/07/2002	07/10/2007	63	0.002	Yes	Increase
		Q4	15	14	11/07/2002	07/10/2007	56	0.005	Yes	Increase
	13	Q1	8	8	08/23/2002	07/23/2007	20	0.007	Yes	Increase
0.40		Q2	8	8	08/23/2002	07/23/2007	8	0.199	No	N/A
		Q4	8	7	08/23/2002	07/23/2007	10	0.138	No	N/A
	19	Q1	17	16	08/22/2002	07/11/2007	53	0.020	Yes	Increase
0.50		Q2	17	14	08/22/2002	07/11/2007	67	0.005	Yes	Increase
		Q3	17	14	08/22/2002	07/11/2007	55	0.017	Yes	Increase
		Q4	17	16	08/22/2002	07/11/2007	66	0.005	Yes	Increase
		Q1	18	18	08/22/2002	07/11/2007	90	0.001	Yes	Increase
0.54	9	Q2	18	18	08/22/2002	07/11/2007	67	0.009	Yes	Increase
		Q3	18	18	08/22/2002	07/11/2007	85	0.001	Yes	Increase
		Q4	18	17	08/22/2002	07/11/2007	74	0.004	Yes	Increase
	18	Q1	19	19	08/23/2002	07/12/2007	89	0.002	Yes	Increase
0.60		Q2	19	18	08/23/2002	07/12/2007	74	0.008	Yes	Increase
0.00		Q3	19	19	08/23/2002	07/12/2007	94	0.001	Yes	Increase
		Q4	19	19	08/23/2002	07/12/2007	95	0.001	Yes	Increase
	3	Q1	19	16	08/23/2002	06/27/2007	44	0.064	No	N/A
		Q2	19	17	08/23/2002	06/27/2007	89	0.002	Yes	Increase
0.75		Q3	19	18	08/23/2002	06/27/2007	66	0.015	Yes	Increase
		Q4	19	18	08/23/2002	06/27/2007	108	0.000	Yes	Increase
	10	Q1	7	5	08/22/2002	07/12/2007	6	0.236	No	N/A
		Q2	7	2	08/22/2002	07/12/2007	7	0.191	No	N/A
		Q3	7	3	08/22/2002	07/12/2007	12	0.052	No	N/A
		Q4	7	2	08/22/2002	07/12/2007	7	0.191	No	N/A
0.79	103	Q1	6	2	03/28/2005	07/12/2007	7	0.136	No	N/A
		Q2	6	2	03/28/2005	07/12/2007	1	0.500	No	N/A
		Q3	6	2	03/28/2005	07/12/2007	5	0.235	No	N/A
		Q4	6	4	03/28/2005	07/12/2007	6	0.186	No	N/A
	7	Q1	19	19	08/23/2002	07/12/2007	9	0.192	No	N/A
0.80		Q2	19	16	08/23/2002	07/12/2007	58	0.027	Yes	Increase
0.80		Q3	19	15	08/23/2002	07/12/2007	46	0.057	No	N/A
		Q4	19	14	08/23/2002	07/12/2007	85	0.002	Yes	Increase
	104	Q1	6	4	03/28/2005	06/20/2007	-4	0.298	No	N/A
1.00		Q2	6	4	03/28/2005	06/20/2007	9	0.068	No	N/A
		Q4	6	2	03/28/2005	06/20/2007	1	0.500	No	N/A

Notes:

- 1 Properties are ordered as a function of increasing distance from the smelter.
- 2 Trend tests were not conducted for properties with fewer than four rounds of sampling, or for properties that were not sampled during round 25.
- 3 All censored (nondetect) measurements were set equal to a concentration slightly lower than the minimum detected value.
- 4 Monotonic trends are significant for probabilities less than or equal to 0.05; significant negative values for the Mann-Kendall test statistic indicate that trends are decreasing; and significant positive values for the Mann-Kendall test statistic indicate that trends are increasing.

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TABLE 2

RESULTS OF LINEAR REGRESSION ANALYSIS FOR ALL QUADRANTS SHOWING A SIGNIFICANT INCREASING MANN-KENDALL TREND TEST RESULT

Distance From Smelter	House		Number of Sampling	Regression Coefficients for Days Versus Concentration			Monthly Increase	95 Percent Confidence Limits for Monthly Increase in Lead Concentrations		Percentiles for the Distribution of Estimated UCLs within Each Distance Group		
(Miles)	Number	Quadrant	Events	Intercept	Slope	S.E. (Slope)	(mg/kg/month)	LCL	UCL	50	75	90
	76	Q1	12	53.79	0.14	0.03	4.35	2.45	6.25			
		Q2	12	94.54	0.09	0.06	2.79	-1.07	6.65			
	5	Q1	18	22.49	0.13	0.02	3.98	2.91	5.05	4.25	4.88	7.64
		Q2	18	34.72	0.11	0.02	3.32	2.36	4.28			
		Q3	18	75.25	0.09	0.02	2.65	1.62	3.68			
		Q4	18	20.71	0.28	0.05	8.37	5.16	11.59			
	6	Q1	18	131.66	0.02	0.03	0.69	-1.05	2.44			
Less than		Q2	18	109.32	0.05	0.03	1.52	-0.18	3.23			
or Equal		Q4	18	86.59	0.02	0.02	0.66	-0.37	1.68			
to 0.25	22	Q1	17	97.04	0.08	0.03	2.30	0.35	4.25			
		Q2	17	189.45	0.10	0.03	3.01	1.31	4.71			
		Q3	17	66.84	0.10	0.02	2.95	1.76	4.13			
		Q4	17	72.54	0.09	0.02	2.74	1.49	3.99			
	24	Q1	15	111.04	0.10	0.02	3.08	1.55	4.62			
		Q2	15	42.03	0.11	0.02	3.36	2.13	4.59			
		Q3	15	55.21	0.06	0.02	1.76	0.69	2.84			
		Q4	15	70.62	0.04	0.02	1.33	0.28	2.38			
0.25 to 0.50	13	Q1	8	146.64	0.12	0.02	3.53	2.03	5.04			
	19	Q1	17	61.48	0.03	0.01	0.83	0.10	1.57	2.27	3.72	5.04
		Q2	17	45.51	0.06	0.01	1.68	0.95	2.40			
		Q3	17	47.05	0.03	0.02	0.81	-0.18	1.81			
		Q4	17	66.78	0.04	0.02	1.23	0.20	2.27			
0.50 to 0.75	9	Q1	18	68.31	0.04	0.01	1.28	0.59	1.96	1.96	3.40	9.03
		Q2	18	75.84	0.06	0.02	1.71	0.37	3.05			
		Q3	18	111.85	0.22	0.06	6.45	2.70	10.21			
		Q4	18	92.07	0.09	0.03	2.66	0.99	4.33			
	18	Q1	19	60.98	0.08	0.02	2.32	1.24	3.40			
		Q2	19	54.35	0.05	0.01	1.45	0.73	2.17			
		Q3	19	70.38	0.03	0.01	0.90	0.45	1.36			
		Q4	19	63.17	0.04	0.01	1.34	0.82	1.87			
		Q2	19	54.12	0.04	0.01	1.16	0.50	1.82			
	3	Q3	19	43.09	0.03	0.01	1.04	0.47	1.61			
		Q4	19	47.04	0.04	0.01	1.19	0.73	1.66			

Notes:

Houses within each distance group are sorted by increasing distance from the smelter

kg Kilogram

LCL Lower confidence limit

mg Milligram

MLE Maximum likelihood estimation

ND Nondetect

OLS Ordinary least squares

S.E. Standard error of estimate

UCL Upper confidence limit

OLS regression was used for cases where all results were detected. Censored MLE regression was used in all cases where one or more measurements were reported as below the detection limit (that is, "ND") following Helsel (2005). All analyses were performed on the data in original units.

Helsel, D. 2005. Nondetects and Data Analysis: Statistics for Censored Environmental Data. John Wiley & Sons, Inc., New York, NY. 250 pages.

FIGURE 1
LEAD CONCENTRATION TRENDS FROM ROUND 7 THROUGH 25



















